

## PATENT SPECIFICATION

(11) 1 514 239

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- (21) Application No. 48196/75 (22) Filed 24 Nov. 1975  
 (23) Complete Specification filed 7 Dec. 1976  
 (44) Complete Specification published 14 June 1978  
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 (52) Index at acceptance C1H 120 414 620 710 726 742 767  
 (72) Inventors AMALENDU JYOTI MAJUMDAR and ROBERT CLAUDE de JEKEY

(54) IMPROVEMENTS IN AND RELATING TO  
FIBRE-REINFORCED COMPOSITES

(71) We, NATIONAL RESEARCH DEVELOPMENT CORPORATION, a British Corporation established by Statute, of Kingsgate House, 66-74 Victoria Street, London, S.W.1, do hereby declare the invention for which we pray that a patent may be granted to us, and the method by which it is

tion also includes a process for making these latter materials including in the general process described above the step of impregnating the material with a monomer preferably before the material is fully cured, and thereafter polymerising the monomer.

The monomer may be an organic monomer and it may contain a catalyst. Polymerisation

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## ERRATUM

SPECIFICATION No. 1,514,239

Page 1, Heading, (72) Inventors, for JEKEY  
*read VEKEY*  
 THE PATENT OFFICE  
*29th August, 1978*

30 a small proportion ---  
 from the flue gases of power stations using  
 pulverised coal as fuel. This ash is usually  
 referred to as "pulverised fuel ash" (P.F.A.).  
 Cenospheres are currently available under the  
 tradename "Fillite".

men heat treated --- to effect  
 polymerisation. Typically methyl methacrylate could be used as the monomer with benzoyl peroxide employed as the catalyst in  
 the conventional way.

35 The cenosphere content of materials according  
 to the present invention is advantageously  
 not in excess of fifty five per cent (by weight)  
 of the other solids present.

The percentage weights of the dry components in eight particular boards produced  
 in accordance with this method are given in  
 the table below. Of these boards 4 to 8 are  
 polymer impregnated, whilst boards 1 to 3  
 are not.

40 In some embodiments of the invention, the  
 material is polymer-impregnated. The inven-

SEE ERRATA CLIP ATTACHED

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(54) IMPROVEMENTS IN AND RELATING TO  
FIBRE-REINFORCED COMPOSITES

5 (71) We, NATIONAL RESEARCH DEVELOPMENT CORPORATION, a British Corporation established by Statute, of Kingsgate House, 66-74 Victoria Street, London, S.W.1, do hereby declare the invention for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

10 The present invention relates to fibre-reinforced composites.

According to the present invention a fibre-reinforced composite comprises a glass-reinforced cement/cenospheres material in which the cenosphere content is not less than thirty per cent by weight of the other solids present.

15 The invention further includes a process for making the composite, including the steps of spraying on to a mould surface reinforcing fibres, a cement slurry, and an amount of cenospheres equal to at least thirty per cent of the weight of the other solids present, partially dewatering the cement slurry e.g. by applying suction to the said surface, and thereafter curing the cement.

20 By "cenospheres" in this specification is meant the minute hollow spheres that form a small proportion of the ash precipitated out from the flue gases of power stations using pulverised coal as fuel. This ash is usually referred to as "pulverised fuel ash" (P.F.A.). Cenospheres are currently available under the tradename "Fillite".

25 The cenosphere content of materials according to the present invention is advantageously not in excess of fifty five per cent (by weight) of the other solids present.

30 In some embodiments of the invention, the material is polymer-impregnated. The inven-

tion also includes a process for making these latter materials including in the general process described above the step of impregnating the material with a monomer preferably before the material is fully cured, and thereafter polymerising the monomer.

35 The monomer may be an organic monomer and it may contain a catalyst. Polymerisation may be effected by heat curing.

The present invention also includes products made from the fibre-reinforced composite of the invention.

40 In particular examples of the invention, ordinary Portland cement (O.P.C.), cenospheres, and water, are mixed with a water/solids ratio of about 3:5. This mix and "Cem-fil" glass fibres are then sprayed on to a vacuum bed mould surface and is dewatered in the usual way by the application of suction to the mould surface. "Cem-fil" is a Trade Mark.

45 The resulting sheet of material can then be removed from the mould surface e.g. using a suction pad, and is left to cure and dry in the usual way. However, if polymer-impregnation is desired then when the sheet of material has partially cured but thoroughly dried it is immersed in an organic monomer containing a catalyst and this monomer is then heat treated in the usual way to effect polymerisation. Typically methyl methacrylate could be used as the monomer with benzoyl peroxide employed as the catalyst in the conventional way.

50 The percentage weights of the dry components in eight particular boards produced in accordance with this method are given in the table below. Of these boards 4 to 8 are polymer impregnated, whilst boards 1 to 3 are not.

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| Boards | OPC  | Cenospheres | Glass fibre | Polymer |
|--------|------|-------------|-------------|---------|
| 1      | 54.6 | 36.0        | 9.4         | —       |
| 2      | 45.6 | 44.2        | 10.2        | —       |
| 3      | 58.4 | 31.4        | 10.2        | —       |
| 4      | 47.1 | 31.0        | 8.1         | 13.7    |
| 5      | 37.4 | 36.3        | 8.4         | 18.0    |
| 6      | 27.0 | 47.0        | 9.5         | 16.5    |
| 7      | 42.9 | 35.1        | 8.0         | 14.0    |
| 8      | 43.5 | 35.5        | 8.1         | 12.9    |

The measured properties of these boards (after 28 days) were as follows:-

| Boards | Density<br>(t/m <sup>3</sup> ) | MOR<br>(MN/m <sup>2</sup> ) | Specific<br>flexural strength<br>(MNmm/t) | Tensile<br>Strength<br>(MN/m <sup>2</sup> ) | Impact<br>Strength<br>(kJ/m <sup>2</sup> ) | Young<br>Modulus<br>(GN/m <sup>2</sup> ) |
|--------|--------------------------------|-----------------------------|---|---|--|--|
| 1      | 1.06                           | 14.1                        | 13.3                                      | 5.5   | 16.1                                       | 9.0                                      |
| 2      | 0.90                           | 10.1                        | 11.2                                      | 4.1   | 11.7                                       | 7.5                                      |
| 3      | 1.07                           | 14.6                        | 13.6                                      | 6.8   | 14.0                                       | 9.8                                      |
| 4      | 1.12                           | 30.8                        | 27.5                                      | 9.8   | 25.9                                       | 11.6                                     |
| 5      | 0.98                           | 22.3                        | 22.6                                      | 10.4  | 21.1                                       | 9.9                                      |
| 6      | 0.84                           | 19.5                        | 23.2                                      | 7.6   | 19.5                                       | 6.4                                      |
| 7      | 1.01                           | 20.5                        | 20.3                                      | —   | 19.3                                       | —  |
| 8      | 1.01                           | 21.2                        | 21.0                                      | —   | 16.0                                       | —  |

The abbreviation MOR stands for modulus of rupture.

5 Where present, polymerisation was effected by heat curing the impregnated sheet for four to twenty hours at a temperature of 60°—80°C in a conventional curing oven (Boards 4 to 7) or in an 80°C hot water bath (Board 8). In all these cases, the sheet was preferably wrapped during curing to

prevent excessive monomer loss by volatilisation.

The results of durability tests conducted on boards 4, 5 and 6 are given below. In the table, the air storage condition refers to air with a relative humidity of 60% at 20°C and the water storage condition refers to complete immersion of the object under water.

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|         |      | Impact Strength (kJ/m <sup>2</sup> ) |         |      |
|---------|------|--------------------------------------|---------|------|
| As made |      | 90 day                               | 180 day | 1 yr |
| 25.9    | 22.9 |                                      | 22.5    |      |
|         | 20.8 |                                      | 19.6    |      |
| 21.1    |      |                                      | 22.4    |      |
|         |      | 20.4                                 | 18.3    |      |
| 19.5    |      | 17.3                                 | 14.2    |      |
|         |      |                                      | 18.6    |      |
| 16.0    |      |                                      | 16.0    |      |
|         |      |                                      | 18.5    |      |
| 14.6    |      |                                      | 14.6    |      |
|         |      |                                      | 15.1    |      |
|         |      |                                      |         | 15.8 |

|   |                       | MOR (MN/m <sup>2</sup> ) |        |         |      |
|---|-----------------------|--------------------------|--------|---------|------|
|   | Storage<br>Conditions | As<br>made               | 90 day | 180 day | 1 yr |
| 4 | Air                   | 30.8                     | 29.4   |         | 26.9 |
|   | Water                 |                          | 27.2   |         | 26.2 |
| 5 | Natural<br>weathering |                          |        |         | 29.8 |
|   | Air                   | 22.3                     |        | 29.1    | 27.3 |
| 6 | Water                 |                          |        | 25.3    | 23.6 |
|   | Natural<br>weathering |                          |        |         | 25.0 |
| 5 | Air                   | 19.5                     |        | 18.5    | 18.7 |
|   | Water                 |                          |        | 15.1    | 18.0 |
| 6 | Natural<br>weathering |                          |        |         | 18.8 |
|   |                       |                          |        |         |      |

As compared with conventional glass-reinforced cement (GRC) materials, the also have the advantages of higher specific flexural strength, improved insulation and

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ducane and as a consequence are expected  
to have a superior fire performance (verified  
in the case of the polymer-impregnated  
materials). Moreover because of their use of  
a non-energy-consuming readily-available by-  
product (cocospheres), the materials of the  
present invention may be cheaper to manu-  
facture than the materials of the prior art.  
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By specific nuclear strength in this con-  
text is meant the flexural strength of the  
material per unit density. As indicated in the  
second table, specific strengths in excess of  
20 meganewton-metres per metric ton have  
been measured for polymer-impregnated com-  
posites according to the present invention.

As compared with available wood-chipboard materials, the polymer-impregnated composites of the present invention have the

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advantages of having a higher specific flexural strength and fire resistance, and of being virtually rot-proof.

As compared with previously proposed GRC/PFA materials, the polymer-impregnated composites of the present invention have the advantage of being lighter and more easily fabricated. They may also prove to be cheaper.

One application for the polymer-impregnated composites of the present invention is as a wood substitute e.g. for flooring panelling. They might also prove to be useful for exposed applications.

One application for the non-impregnated composites of the present invention might be for wall boards and in this context they have the advantage of having a higher impact strength than many conventional insulation materials.

#### WHAT WE CLAIM IS:—

1. A process for making a fibre-reinforced composite, including the steps of spraying on to a mould surface reinforcing fibres, a cement slurry, and an amount of cenospheres (as hereinafter defined) equal to at least thirty per cent of the weight of the other solids present, partially dewatering the cement slurry, and thereafter curing the cement.

2. A process as claimed in Claim 1 in which the cenosphere content does not exceed fifty-five per cent (by weight) of the other solids present.

3. A process as claimed in Claim 1 or 2 including the steps of impregnating the material with a monomer and thereafter polymerising the monomer.

4. A process as claimed in Claim 3 in which the step of impregnating the material with a monomer is carried out before the material is fully cured.

5. A process as claimed in Claim 3 or Claim 4 in which the monomer is an organic monomer.

6. A process as claimed in Claim 5 in which the monomer contains a catalyst.

7. A process as claimed in any of Claims 3 to 6 in which polymerisation is effected by heat curing.

8. A fibre-reinforced composite comprising glass-reinforced cement cenospheres material in which the cenosphere content is not less than thirty per cent by weight of the other solids present.

9. A fibre-reinforced composite as claimed in Claim 8 in which the cenosphere content does not exceed fifty-five per cent by weight of the other solids present.

10. A fibre-reinforced composite as claimed in Claim 8 or Claim 9 for use as a wall board or like heat insulating component.

11. A fibre-reinforced composite as claimed in any of Claims 8 to 10 when prepared by a process according to Claim 1 or Claim 2.

12. A fibre-reinforced composite as claimed in any of Claims 8 to 11 when prepared by a process according to any of Claims 3 to 7.

13. A fibre-reinforced composite as claimed in Claim 12 for use as a wood substitute.

14. A fibre-reinforced composite as claimed in Claim 13 having a specific strength in excess of 20 meganewton-metres per metric ton.

15. A fibre-reinforced composite as claimed in Claim 8 and substantially as hereinbefore described in the examples hereinbefore quoted.

16. A product made from a fibre-reinforced composite according to any of Claims 8 to 15.

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